



Increased dust deposition in the Parece Vela Basin since the mid- Pleistocene inferred from radiogenic Sr and Nd isotopes

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ABSTRACT

Eolian dust deposition is crucial to paleoclimate reconstructions of dust source areas and may have played an important role in the climate during the mid-Pleistocene transition (MPT). Here, we present the radiogenic Sr and Nd isotopic composition and excess barium (Ba_{xs}) of a sediment core that was recovered from the Parece Vela Basin (PVB) in the northwestern Pacific. The new record indicates that these detrital sediments are primarily composed of dust from western and central Chinese deserts and volcanic materials from the islands in and around the PVB. A two end-member Sr isotope model was applied to distinguish Asian dust from volcanic materials and to reconstruct the mass accumulation rates (MARs) of dust over the last 2.1 Myr. Starting at ~ 1.2 Myr, the dust MARs doubled relative to those in the Early Pleistocene, which was coupled with the frequent congruence of minimum eccentricity and low obliquity. We argue that the congruent minimum eccentricity and low obliquity may have triggered the climate change before and during the MPT and resulted in the aridity of East Asia and increased dust input. The coupling of dust MAR, Ba_{xs} MAR and atmospheric CO_2 concentration and their rapid increase since the MPT strongly suggest that the internal positive feedback of dust input might be another mechanism behind dust input to the PVB since the MPT.

1. Introduction

Eolian dust that is derived from arid continental areas is widely distributed in the atmosphere, seawater and ocean floor sediments. Atmospherically transported eolian dust may play a critical role in the Earth's climate system by influencing the radiative balance of the atmosphere (Tegen et al., 1996). Dust that falls into the ocean potentially influences the structure of oceanic ecosystems and biogeochemical cycles, thus influencing the partitioning of CO_2 among the atmosphere, deep sea and climate (e.g., Martin, 1990; Watson et al., 2000). In addition, dust deposition on the deep ocean floor can reveal important clues about past climate conditions of the dust source area (e.g., Rea, 1994; Rea et al., 1998; Pettke et al., 2000). The temporal variability of dust mass accumulation rates (MARs) provides a signature of the aridity history of the dust's source regions (Rea, 1994). Furthermore, dust MARs can be used to evaluate the role of dust in climate change (e.g., Jickells et al., 2005; Martínez-García et al., 2011). Therefore,

substantial efforts have been made to reconstruct the spatial and temporal variability of dust MARs in the ocean on different time scales (Clemens et al., 1996; Rea et al., 1998). The northwestern Pacific is the most important sink area for dust that is transported from the Asian continent (e.g., Mahoney, 2005; Jiang et al., 2013a, 2016; Seo et al., 2014). The Parece Vela Basin (PVB) is a back-arc basin behind the Izu-Bonin-Mariana arc systems in the northwestern Pacific (Fig. 1). Clay minerals, grain sizes and radiogenic Sr and Nd isotopic evidence have demonstrated that Asian dust is a major component of the PVB (Ming et al., 2014; Seo et al., 2014; Zhou et al., 2015). Similarly, variations in the dust signals have been correlated to climate change in East Asia (Asahara, 1999). Dust delivery to the ocean can fertilize the marine plankton by supplying essential nutrients (e.g., Fe) and modulate the uptake of carbon and CO_2 concentration in marine ecosystems, further affecting the climate indirectly (Martin, 1990). However, the amount of dust that has been transported to and deposited in this area is not clear. In addition, the widely distributed thickly laminated diatom

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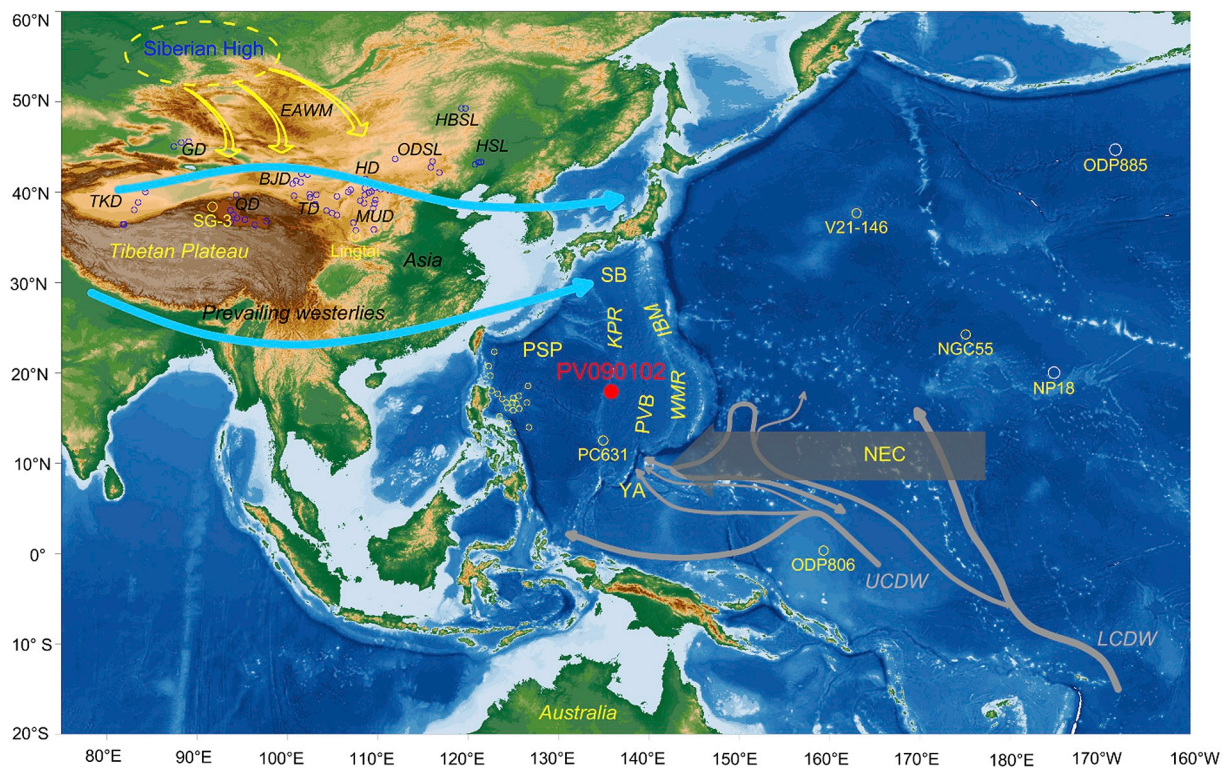


Fig. 1. Map showing the locations of core PV090102 (red dot, this study) and other sediment cores that are discussed in the text: ODP 885A (Rea et al., 1998), V21-146 (Hovan et al., 1991), PC631 (Seo et al., 2014), NGC55, and NP18 (Asahara, 1999) and ODP806 (Patterson et al., 1999) in the North Pacific; SG-3 in the Qaidam Basin (Cai et al., 2012); and the Lingtai section on the Chinese Loess Plateau (Sun and An, 2005). Also shown are the desert provinces of Asia (Chen et al., 2007), including the northern Chinese deserts (NCDs, e.g., G.D., Gurbantunggut Desert; ODSL, Onqin Daga Sandy Land; HBSL, Hunlun Buir Sandy Land; and HSL, Horqin Sandy Land), western and central China deserts (WCCDs, e.g., TKD, Taklimakan Desert; QD, Qaidam Desert; TD, Tengger Desert; and BJD, Badain Jaran Desert), and East China deserts (ECDs, e.g., HD, Hobq Desert; and MUD, Mu Us Desert). The sites analyzed by Chen et al. (2007) are plotted with blue circles. The East Asian winter monsoon (EAWM) wind directions and prevailing westerly trajectories are shown with light blue solid and dashed lines, respectively. The present-day flow paths of the major currents of the North Equatorial Current (NEC) are shown with a dark gray arrow. The light gray arrows show the eastern and western branch currents of deep circulation in the lower deep layer (> 3500 m), which carry Lower Circumpolar Deep Water (LCDW), and the deep-circulation current in the upper deep layer (2000–3500 m), which carries Upper Circumpolar Deep Water (UCDW) that flows along the western edge of the equatorial Pacific and enters the Philippine Sea (Kawabe et al., 2009). PSP, Philippine Sea Plate; PVB, Parece Vela Basin; KPR, Kyushu-Palau Ridge; SB, Shikoku Basin; WMR, Western Mariana Ridge; IBM, Izu-Bonin-Mariana; YA, Yap Arc. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

(*Ethmodiscus rex*) mats in the PVB during the Last Glacial Maximum (Zhai et al., 2009) likely play a significant role in the global carbon and silicon cycles and global climate change (Kemp et al., 2006). Laminated diatom mat accumulation is believed to result from the input of iron and silicon in Asian dust (Xiong et al., 2013, 2015) and well-developed manganese nodules may be related to Asian dust input for similar reasons (Xu et al., 2007). However, no direct evidence demonstrates that eolian dust influences the formation of diatom mats and manganese nodules, and a reasonable link between Asian dust and climate change is not yet available because of the absence of a temporal and spatial record of dust input in this area.

The combination of strontium (Sr) and neodymium (Nd) isotopic signatures has been shown to reliably discriminate between inputs from different detrital sediment source areas (Weldeab et al., 2002; Jiang et al., 2013a, 2013b, 2016). A two end-member model of Sr isotopes can be used to quantitatively separate isotopically different end-members (Faure, 1986). This model has been successfully applied to reconstruct detrital MARs in the Arctic Ocean (Tütken et al., 2002), Okinawa Trough (Meng et al., 2001) and Mediterranean Sea (Weldeab et al., 2002).

The intent of this study is to determine the dust input sources to the PVB based on the Sr and Nd isotopic composition of detrital sediments in a sediment core that was recovered from the PVB and to quantitatively reconstruct the temporal variations in the MARs of eolian dust over the last 2.1 Myr. In order to demonstrate the feedback mechanism

of dust input on the climate, we reconstructed the MAR of excess barium (Ba_{xs}) to indicate the variation in export production in the PVB over the last 2.1 Myr. These results were then used to infer the climatic conditions in the dust source areas and to evaluate the potential mechanisms of these dust variations during the mid-Pleistocene transition (MPT).

2. Geological background

The PVB is located in the southeastern region of the Philippine Sea Plate (Fig. 1). The basin is an elongated, spindle-shaped back-arc basin that is bounded by the West Mariana Ridge to the east, the Kyushu-Palau Ridge to the west, and the Izu-Bonin Mariana and Yap Arcs to the south; the basin merges with the Shikoku Basin to the north at 24° N. The average water depth of the PVB floor exceeds 4000 m (Okino et al., 1998). The topography of the eastern PVB is rougher and shallower than that of the western portion, and the rifting zone is in the middle (Karig, 1975). The sediment in this area is mainly composed of pelagic clays, and manganese nodules are common in the seafloor surface sediments based on the investigation of Deep Sea Drilling Project Leg 59 (Shipboard Scientific Party, 1978). Diatom mats that range in thickness from tens of centimeters to several meters are also found in the area (Shipboard Scientific Party, 1978; Zhai et al., 2009), and these well-preserved diatom mats were believed to have formed during the Last Glacial Maximum (Xiong et al., 2013). The formation of the PVB began

at approximately 25 Myr, and the main spreading stage of the PVB ceased after 17 Myr (Ishizuka et al., 2011).

The oceanic surface current that influences the PVB is the westward-flowing North Equatorial Current (Fig. 1). A minor westward branch bifurcated from Lower Circumpolar Deep Water, and two minor northern branches separated from Upper Circumpolar Deep Water enter the PVB at the southern end of Izu-Bonin Mariana arc system (Kawabe et al., 2009).

Eolian dust from the Asian continent (primarily deserts, sandy areas and loess plateau) have three potential source regions (Chen et al., 2007): northern Chinese deserts (NCDs), which include the Gurbantunggut Desert, Onqin Daga Sandy Land, Hunlun Buir Sandy Land, and Horqin Sandy Land; western and central Chinese deserts (WCCDs), which include the Taklimakan, Qaidam, Tengger and Badain Jaran Deserts; and eastern Chinese deserts (ECDs), which include the Hobq and Mu Us Deserts (Fig. 1). The main source areas of Asian dust are located within the area influenced by the monsoon and/or westerly circulations. The low-level atmospheric circulation over most of northern China is dominated by the winter monsoon, which is associated with winds blowing out of the Siberian high pressure zone, while high levels are dominated by zonal westerly flow (Fig. 1). The tropical northwest Pacific is located downwind and is influenced by the prevailing westerlies. Asian dust can be transported to the tropical northwestern Pacific from the Asian continent by the prevailing westerlies (Duce et al., 1980; Rea, 1994; Jiang et al., 2016).

3. Materials and methods

The core PV090102 that was used in this study was collected with a gravity sampler from the western part of the PVB (135.878°E, 17.921°N, Fig. 1) at a water depth of 4350 m using the research vessel Ke Xue Yi Hao during a cruise in the northwestern Pacific in 2004. The lithology of the 378 cm-long section is essentially uniform and consists mainly of yellowish-brown and dark-brown pelagic clays. The only fossils are siliceous radiolarians and diatoms, which indicates that this interval was deposited below the carbonate compensation depth. The age model for core PV090102 was established according to paleo-magnetic chronology (Fig. 2) (Meng et al., 2010). The depositional age at the bottom of the core is estimated to be approximately 2.1 Myr, and the linear sedimentation rate (LSR) averages 2–3 mm/kyr, which is similar to other sections in the North Pacific (Rea, 1994). The low sedimentation rates and 8 cm sampling interval yield a temporal resolution of approximately 45 kyr per sample. After the core was split, the wet bulk sediments were placed in containers and then dried and weighed. The dry bulk density (DBD) was directly calculated from the mass (weight) of the dry bulk sediment sample divided by the total volume of the wet sample.

A total of 47 samples from the core were collected for Sr and Nd isotope analyses (Fig. 1 and Table S1). Approximately 400 mg of bulk sediment from each sample was dried at 50 °C and homogenized in an agate mortar. To determine the pure detrital radiogenic Sr and Nd isotope signatures, we used the leaching procedure that was described in Gutjahr et al. (2007) and Bayon et al. (2002). Sample splits of 300–400 mg each were weighed precisely (on a balance with 0.1 mg precision) and then treated with distilled water, NaOH (1 M), hydroxylamine hydrochloride (0.05 M), and hydrogen peroxide (5%) at room temperature to remove sea salts, diatoms, ferromanganese coatings, and organic matter, respectively. Subsequently, the residual detrital fractions were dried at 50 °C, weighed precisely and ground again, and then their Sr and Nd isotopic signatures were determined. The difference in weight before and after the leaching procedure is considered the weight of the non-detrital fraction, which was used to calculate the detrital fraction (DF) content of the bulk sediment.

Approximately 100 mg of each detrital sample was used to analyze the Sr and Nd isotopic compositions. Each sample was totally dissolved in an airtight Teflon bomb with a mixture of concentrated HClO₄, HNO₃

and HF and heated on a hot plate (at 120 °C) for seven days. Rb and Sr were separated using Bio-Rad AG50W-X12 and a 200–400 mesh cation exchange resin. 6 N HCl was used to pre-clean the column, and 2.5 N HCl was then used to pre-condition the column. After loading the samples, 2.5 N HCl was added to rinse, elute and collect Sr; then, 6 N HCl was used to elute and collect REE. The collected REE solution was used to separate Sm and Nd using P507 cation exchange resin. 6 N HCl and 0.08 N HCl were used to pre-clean and pre-condition the column, and 0.15 N HCl was used to elute and collect Nd. Radiogenic Sr and Nd isotope measurements were performed on a Nu Plasma HRMC-ICP-MS (High-Resolution Multicollector Inductively Coupled Plasma Mass Spectrometer) instrument at the Key Laboratory of Marine Sedimentology and Environmental Geology, First Institute of Oceanography, State Oceanic Administration, China. The ⁸⁷Sr/⁸⁶Sr result that was measured for the reference material NBS 987 was 0.710340 ± 3 (2σ , $n = 10$), and its reference value was 0.710341 ± 26 . The ¹⁴³Nd/¹⁴⁴Nd result that was measured for the reference material JNdi-1 was 0.512117 ± 9 (2σ , $n = 10$), and its reference value was 0.512115 (Tanaka et al., 2000). Procedural blanks for this processing technique were approximately 100 picograms for Sr and 15 picograms for Nd. Complete duplicate analyses of some samples show that the reproducibility of the Sr and Nd isotopic measurements is better than 100 ppm (Table S1). The ¹⁴³Nd/¹⁴⁴Nd ratios were normalized to the value for the Chondritic Uniform Reservoir (CHUR) and reported as $\epsilon_{Nd}(0) = ((^{143}\text{Nd}/^{144}\text{Nd})_{\text{sample}} / 0.512630) - 1 \times 10^4$ (Bouvier et al., 2008).

To obtain biogenic barium (Ba), termed as excess barium (Ba_{xs}), and to evaluate the relationship between the variation in paleo-productivity and the dust MARs in the PVB, the concentrations of three elements Al₂O₃, TiO₂ and Ba—of 47 sediment samples from core PV090102 were determined with X-ray fluorescence spectrometry at the Institute of Geophysical and Geochemical Exploration, Chinese Academy of Geological Sciences. The detection limits of the three elements were 0.2 wt%, 0.05 wt% and 5 µg/g, respectively. Duplicate samples and a suite of rock and sediment standard samples (GAU2GSS1, GAU9GSS3, GAU10GSD9 and GAU11GSD10) were also measured to monitor instrument precision and assess data quality. The analytical precision (RSD) for all the three elements was better than 5%.

Ti was used to normalize Ba concentrations in order to avoid dilution effects (Lyons and Berner, 1992; Xiong et al., 2012). Ba_{xs} was calculated as the total Ba concentration minus the estimated detrital Ba contribution ($\text{Ba}_{\text{xs}} = \text{Ba}_{\text{sample}} - \text{Ti}_{\text{sample}} \times (\text{Ba}/\text{Ti})_{\text{PAAS}}$) (Dymond et al., 1992; François et al., 1995; Xiong et al., 2012). Here, the Ba/Ti ratio of Post-Archean Australian Shale (PAAS) (Taylor and McLennan, 1985) was used for normalization. The MAR of Ba_{xs} was obtained from the concentration of Ba_{xs} multiplied by SR and DBD.

4. Results

The Sr and Nd isotope values for the detrital fraction of the samples from core PV090102 are presented in Table S1 and Fig. 3. The Sr isotopic variation of two core sediments from Asahara (1999) are also displayed in Fig. 3 for comparison. The ⁸⁷Sr/⁸⁶Sr values of the detrital sediment in site PV090102 (varied from 0.70956 to 0.71339) are similar to those of sediments in the central North Pacific (Pettke et al., 2000) but significantly more evolved than those of volcanic rock in the northwestern Pacific (Cohen and O'Nions, 1982; Woodhead, 1989; Volpe et al., 1987, 1990; Hickey-Vargas, 1991). Although the Sr isotope of PV090102 is overall less evolved than the sediments in the northwest Pacific Basin (e.g., sties NGC55 and NP18, Fig. 1) (Asahara, 1999), their down-core variation in the Sr and Nd isotopes is similar (Fig. 3). Three stages can be observed in PV090102: (1) From 2.1 Myr to 1.5 Myr, the sediment had the lowest radiogenic Sr values (average 0.71081) and the highest radiogenic Nd values (average = 7.4). (2) From 1.5–0.6 Myr, the ⁸⁷Sr/⁸⁶Sr values increased gradually (average 0.71232), whereas the $\epsilon_{Nd}(0)$ values decreased (average = 8.6). The

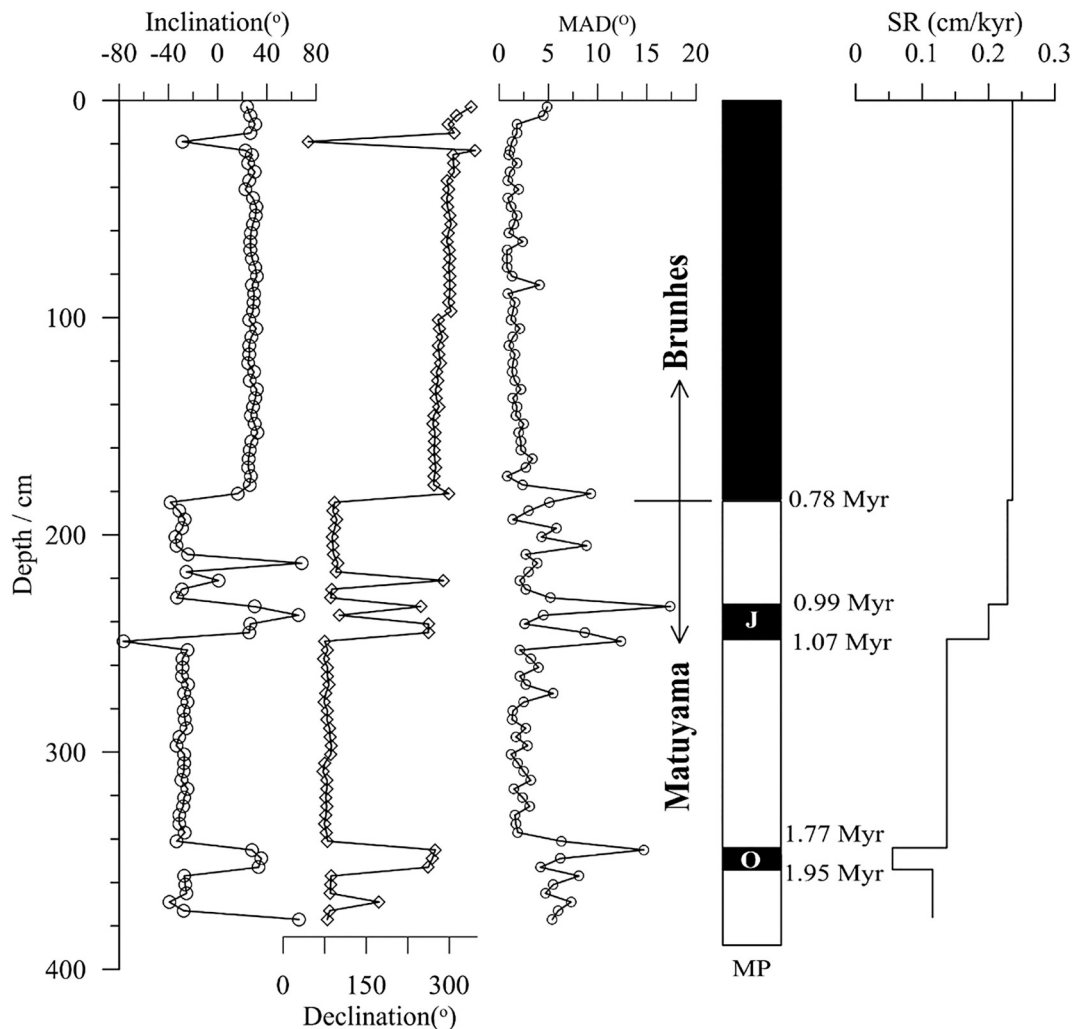


Fig. 2. Magnetostratigraphy and sedimentary rate (SR) of the core PV090102 sediments (the inclination, declination and MAD data are adopted from Meng et al., 2010; MAD-maximum angular deviation; MP-magnetic polarity; J- Jaramillo subchron and O-Olduvai subchron).

most evolved Sr and Nd isotopes were observed within the time interval 1.2–0.9 Myr, with an average of 0.71275 for $^{87}\text{Sr}/^{86}\text{Sr}$ and -8.6 for $\epsilon_{\text{Nd}}(0)$. (3) From 0.6 Myr to the present, the Sr and Nd isotopic compositions became less radiogenic, with an average of 0.71112 for $^{87}\text{Sr}/^{86}\text{Sr}$ and -8.0 for $\epsilon_{\text{Nd}}(0)$.

The concentrations of Al_2O_3 , TiO_2 , Ba and Al/Ti are displayed in Table S2. The Ba/Ti ratio, Ba_{xs} concentration and MAR are displayed in Table S2 and Fig. 3. The Ba_{xs} concentration ranged from 2105.9 to 3798.1 $\mu\text{g/g}$, with an average of 2796 $\mu\text{g/g}$, which is significantly more abundant than that in the upper continental crust (UCC, 550 $\mu\text{g/g}$) and PAAS (650 $\mu\text{g/g}$) (Taylor and McLennan, 1985) and the volcanic rock in and around the PVB (averaged 83.9 $\mu\text{g/g}$, $n = 252$) (Bougault et al., 1982; Sharaskin, 1982; Matthey et al., 1980; Wood et al., 1980; Hawkins et al., 1990). The variation in the Al/Ti ratio ranged from 16.4 to 19.6, with an average of 18.4. The Al/Ti ratios were similar to those reported by Xiong et al. (2012) (average 17.7) and PAAS (16.8). The Ba/Ti ratios varied from 0.15 to 0.30, with an average of 0.21. High Ba/Ti ratios (average 0.24) were observed during 0.6–1.5 Myr, and the highest Ba/Ti ratio (0.29) was observed at approximately 1.2 Myr. Obviously, Ba is enriched in the PVB sediment and more abundant than in the UCC (0.206) and PAAS (0.108). Xiong et al. (2012) argue that the high Ba/Ti ratios indicate that the excess Ba in the PVB results from biological processes rather than originating from opal and ferromanganese oxyhydroxides (Xiong et al., 2012). The estimated Ba_{xs} displayed similar variation to that of the Ba/Ti ratio, showing high concentration during

the time interval of 0.6–1.5 Myr (average 2739 $\mu\text{g/g}$), and the highest Ba_{xs} (3299 $\mu\text{g/g}$) was observed at 1.2 Myr. The Ba_{xs} MAR varied from 47.5 to 318.1 $\mu\text{g cm}^{-2} \text{kyr}^{-1}$, with an average of 220.2 $\mu\text{g cm}^{-2} \text{kyr}^{-1}$ (Table S2). The Ba_{xs} MAR gradually increased starting 1.5 Myr, and a rapid increase occurred at 1.2 Myr (Fig. 3). The overall MAR of Ba_{xs} after MPT was significantly higher than before. The maximum Ba_{xs} MAR occurred at 0.9–1.0 Myr.

5. Discussion

5.1. Potential mixing of sediment end-members

The Sr and Nd isotope signatures of the sediments in the marginal basins of the western Pacific are intermediate between older continental signatures and younger volcanic arc signatures and are more variable (Asahara et al., 1995; Mahoney, 2005; Jiang et al., 2013a). Previous studies have shown that old materials from the Asian continent (Sun, 2005; Chen et al., 2007) and eastern Taiwan orogen (Bentahila et al., 2008) have more evolved Sr and Nd isotopic compositions ($^{87}\text{Sr}/^{86}\text{Sr}$ ratios > 0.71000 and $\epsilon_{\text{Nd}}(0) < -9$), whereas materials from the relatively young crustal provinces around the Pacific Rim, such as from the Izu-Bonin Mariana arc (e.g., Volpe et al., 1987, 1990; Bloomer and Hawkins, 1987; Gribble et al., 1996, 1998) and Luzon Islands (Defant et al., 1990), are characterized by less radiogenic Sr and highly radiogenic Nd isotope compositions (with $^{87}\text{Sr}/^{86}\text{Sr}$ ratios $<$

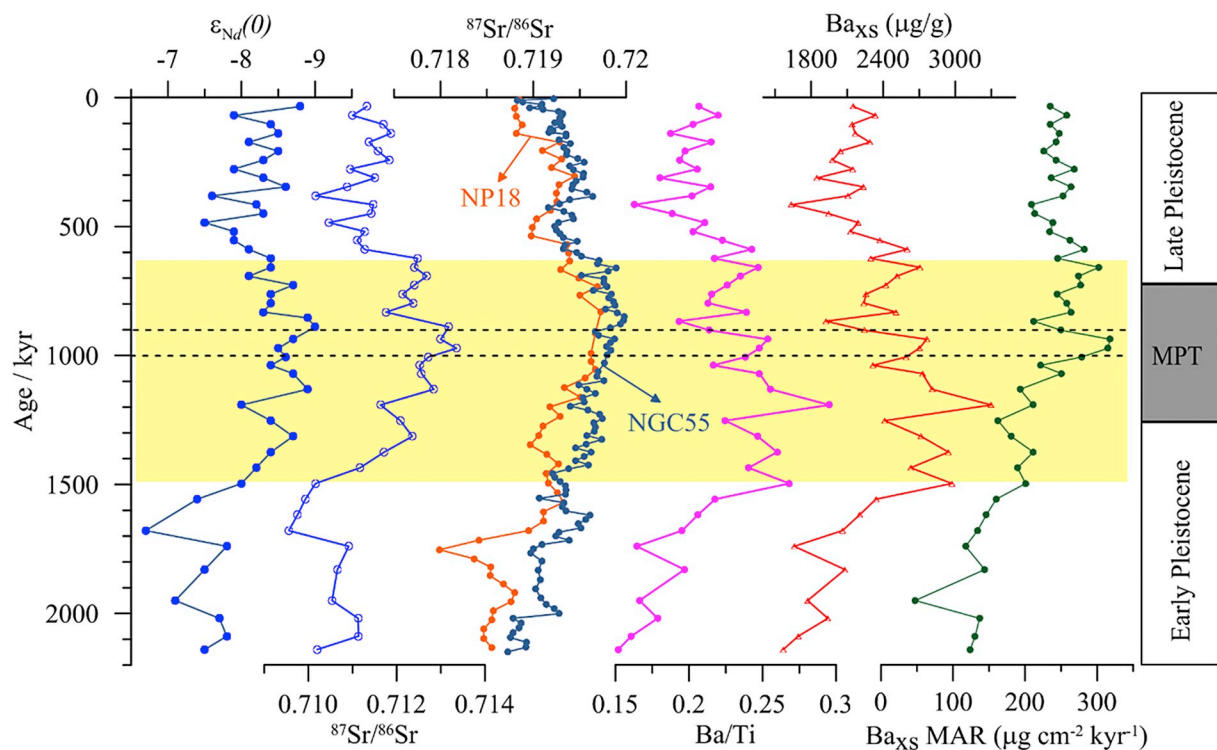


Fig. 3. Down-core variations in the Sr and Nd isotopic signatures, Ba/Ti ratio, excess Ba (Baxs) and Baxs MAR in the detrital fraction of core PV090102 (NP18 and NGC55 are two cores from Asahara, 1999; the yellow bar indicates the more radiogenic Sr isotope at time interval 1.5–0.6 Myr; the dashed lines indicate the highest MAR of Baxs at time interval 1.0–0.9 Myr; the time interval of the MPT is 1.25–0.7 Myr (Clark et al., 2006)). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

0.705 and $\epsilon_{Nd}(0) > +5$). Sediments from southwestern Japan have transitional Nd isotope values ($\epsilon_{Nd}(0) = (-6) - (+4)$) (Nakamura et al., 1990). Although studies have demonstrated that the sediment in the northwestern Pacific represents variable degrees of mixing between old Asian continental dust and juvenile Pacific rim volcanic material (Mahoney, 2005; Jiang et al., 2013a), the potential contributions to the more evolved Sr and Nd isotopes in the PVB sediments demand more detailed analysis.

The published $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ ratios of the sediments and rocks from potential source areas and the detrital fractions in the sediments of core PV090102 are graphically displayed in Fig. 4. The Sr and Nd isotopic signatures of PVB detrital sediments are intermediate between those of volcanic materials from island arcs and dust deposition from the central North Pacific (Pettke et al., 2000) and are different as a whole from those of other potential sediment sources (e.g., Taiwan orogen sediments (Chen et al., 1990; Chen and Lee, 1990) and the Shikoku Basin sediments (Mahoney, 2005)).

The Izu-Bonin-Mariana oceanic arc and Kyushu-Palau Ridge have potentially supplied volcanic ash and volcanoclastic debris (typified by tholeiitic basalt that is characterized by juvenile $\epsilon_{Nd}(0)$ values from $(+8)$ to $(+9)$) to the PVB since the Miocene (Curtis and Echols, 1980). The Mariana arc system became active at 7 Myr, and the Mariana Trough has been spreading since approximately 3.5 Myr (Yamazaki et al., 1993). Volcanic material erupted from the Izu-Bonin-Mariana can be transported westward via the prevailing easterlies (Lee et al., 1995). Therefore, the less radiogenic Sr and more radiogenic Nd were likely derived from explosive volcanism along the active Mariana arc system. In addition, the local weathering of island-arc volcanic rock in and around the PVB may have also contributed juvenile Sr and Nd isotopes transported via local circulation to the PVB.

The Shikoku Basin is just north of the PVB. The sediment in the Shikoku Basin is characterized by evolved Nd isotope values (from (-2) to (-9)) and supplied with juvenile sediment from the Izu-Bonin

arc ($\epsilon_{Nd}(0) = (+8) - (+9)$), transitional sediment from southwestern Japan ($\epsilon_{Nd}(0) = (-6) - (+4)$), and old Asian continent sediment ($\epsilon_{Nd}(0) < (-10)$) (Mahoney, 2005). However, the Nd isotopic signatures of the PVB sediments are different from those of the Shikoku Basin (Fig. 4). The $\epsilon_{Nd}(0)$ values of the PVB are generally lower than those of the Shikoku Basin, although the $^{87}\text{Sr}/^{86}\text{Sr}$ values are the same, which implies that the contribution of Japanese arc material to the study area is negligible.

The Taiwan orogen could be responsible for the evolved Sr and Nd isotope values of the PVB because of its high erosion rate (Dadson et al., 2003) and evolved Sr and Nd isotopic signatures. However, the PVB samples do not fall on the mixing curve between Taiwanese sediment and island arc material (Fig. 4). In addition, previous studies have shown that Taiwanese sediment was mainly trapped in the Huatung Basin (Jiang et al., 2013a) and Okinawa Trough (Bentahila et al., 2008). Therefore, the Taiwan orogen is not a major contributor to the PVB. The radiogenic Sr and Nd isotopic signatures and clay mineralogy indicate that neither Australian nor North African dust contributed a detectable amount to the site of core PC631 in the western Philippine Sea (Seo et al., 2014). Because this area is < 600 km south of core PV090102 (Fig. 1), we argue that the influence of Australian and North African dust on our study site is also negligible and suggest that the Asian continent is the dominant contributor of evolved Sr and Nd isotopes to the PVB. The plot of $^{87}\text{Sr}/^{86}\text{Sr}$ vs $\epsilon_{Nd}(0)$ values for core PV090102's detrital sediment samples illustrates mixing between two isotopically different end-members (Fig. 4), which strongly suggests that the sediment in the PVB is a mixture of Asian dust and juvenile volcanic materials. This conclusion is also supported by the bimodal grain size distribution of the detrital sediment in core PV090102 (Zhou et al., 2015). In addition, the fine grain size (average $3.2\mu\text{m}$) of the detrital fractions, which is $< 16\mu\text{m}$ for the terrigenous input in core PV090102 (Zhou et al., 2015), indicates the negligible effect of hemipelagic and winnowing processes on sedimentation (Rea and Hovan, 1995; Seo

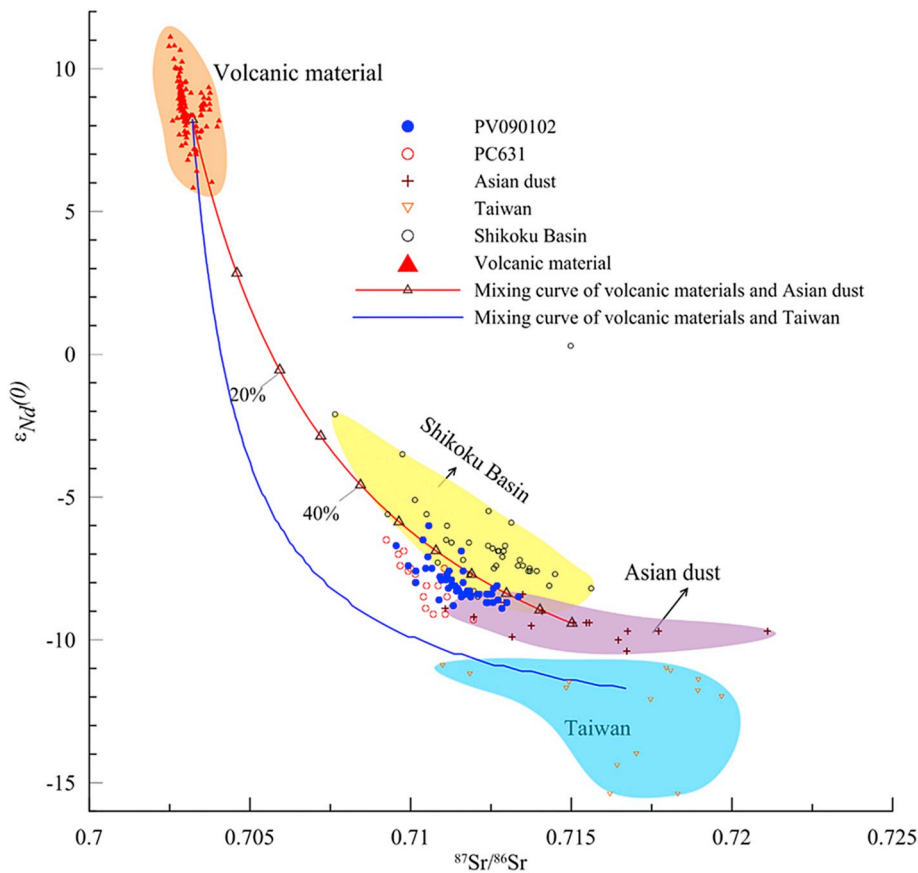


Fig. 4. $^{87}\text{Sr}/^{86}\text{Sr}$ vs $\epsilon_{\text{Nd}}(0)$ values of the detrital fraction of the core PV090102 sediments and potential sediment and rock source areas (literature data). The red line is the mixing curve between volcanic material and Asian dust, which is represented by North Pacific sediments. The relative percentages of Asian dust are marked with empty triangles on the line, with each point indicating 10%. The blue line is the mixing curve between volcanic material and Taiwan orogen sediments and rock. The Sr and Nd isotopic compositions of Asian dust, the Taiwan orogen and Shikoku Basin in this plot are adopted from Pettke et al. (2000), Chen et al. (1990), Chen and Lee (1990), and Mahoney (2005), respectively, and volcanic glass in the Izu-Bonin Mariana arc from the GeoRoc database (Bloomer and Hawkins, 1987; Brounce et al., 2014; Fryer et al., 1982; Gribble et al., 1996, 1998; Hart et al., 1972; Hawkins et al., 1990; Ito and Stern, 1980; Lonsdale and Hawkins, 1985; Matthey et al., 1980; Pearce et al., 2005; Reagan and Meijer, 1984; Sinton and Fryer, 1987; Stern et al., 1991, 2013; Straub, 1995, 1997; Volpe et al., 1987, 1990). The mixing lines in the $^{87}\text{Sr}/^{86}\text{Sr}$ vs $\epsilon_{\text{Nd}}(0)$ plot were developed from known concentrations and isotopic compositions of Sr and Nd from the following areas: Izu-Bonin-Mariana arc system ($\text{Sr} = 183.9$ ppm, $\text{Nd} = 9.96$ ppm, $^{87}\text{Sr}/^{86}\text{Sr} = 0.703208$ and $^{143}\text{Nd}/^{144}\text{Nd} = 0.513052$); Asian dust ($\text{Sr} = 219.1$ ppm, $\text{Nd} = 39.4$ ppm, $^{87}\text{Sr}/^{86}\text{Sr} = 0.71503$ and $^{143}\text{Nd}/^{144}\text{Nd} = 0.512147$, $n = 15$) (Pettke et al., 2000) and the Taiwan orogen ($\text{Sr} = 58$ ppm, $\text{Nd} = 31$ ppm, $^{87}\text{Sr}/^{86}\text{Sr} = 0.71669$ and $^{143}\text{Nd}/^{144}\text{Nd} = 0.51203$) (Chen et al., 1990). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

et al., 2014). The influence of fluvial deposition from the Asian continent is also negligible because of the distance from the Asian continent and the blocking effect of the Kuroshio Current and Ryukyu Islands.

The Sr and Nd isotopic compositions of Asian dust from the three potential source regions (NCDs, ECDs and WCCDs) are different (Fig. 1 and Fig. 5) (Chen et al., 2007; Chen and Li, 2013). The NCDs are characterized by less radiogenic Sr and more radiogenic Nd isotopic signatures, the ECDs are characterized by more radiogenic Sr and less radiogenic Nd isotopic signatures, and the WCCDs are characterized by Sr and Nd isotopic values that are intermediate between those of the NCDs and ECDs. The Nd isotope values show limited overlap between the three regions, which implies highly differentiated material. The Sr and Nd isotopic compositions of most Asian dust in central North Pacific sediments (Pettke et al., 2000) overlap with those of the WCCDs, which indicates that dust deposition to the central North Pacific is mainly derived from the WCCDs. The Nd isotopic composition of Chinese loess (Sun 2005; Wang et al., 2007; Chen and Li, 2013) resembles that of the WCCDs, which suggests an origin in the WCCDs (Sun, 2005; Chen et al., 2007). The Sr isotope values are more radiogenic in Chinese loess than in the WCCDs, supporting the grain size dependence of Sr isotopes (Feng et al., 2009). The NCDs are more radiogenic in Nd than the PVB, which does not support the NCDs as a potential contributor to the PVB (Fig. 5). The Nd isotopic values of the PV090102 detrital fractions do not correspond with the mixing hyperbola between volcanic materials around the PVB and Chinese loess (Fig. 5) and display an offset toward less radiogenic Nd, which indicates that Chinese loess is not a major dust contributor to the PVB. The less radiogenic Nd in the ECDs indicates that these deserts are potential contributors to the PVB (Chen et al., 2007; Seo et al., 2014). However, most of our samples plot close to the mixing curve of volcanic material and WCCDs, which

supports the conclusion that the Asian dust component in the core PV090102 sediments is mainly derived from the WCCDs.

5.2. Increased dust input since the mid-pleistocene

The MARs of dust in sediment core PV090102 are calculated as follows:

$$\text{MAR} (\text{g cm}^{-2}\text{kyr}^{-1}) = \text{LSR} \times \text{DBD} \times \text{DF} \times f_{\text{dust}} \quad (1)$$

where LSR is the linear sedimentation rate (cm kyr^{-1}), DBD is the dry bulk density of the sediment (g cm^{-3}), DF is the weight percentage of the detrital fraction in the bulk sediment, and f_{dust} is the fraction of eolian dust in the detrital sediment.

For a two end-component mixture, a preliminary quantification of the contributions of both end-members to the core sediments can be performed with a two-component mixing model of Sr isotopic signatures (Faure, 1986). The mixing proportions of eolian dust (f_{dust}) were calculated with the following mixing equation developed by Faure (1986):

$$f_{\text{dust}} = \frac{(\text{Sr})_B \times [(X)_B - (X)_M] / \{(\text{Sr})_A \times [(X)_M - (X)_A] + (\text{Sr})_B \times [(X)_B - (X)_M]\}}{1} \quad (2)$$

where $(\text{Sr})_A$ and $(X)_A$ are the concentration and isotopic ratio of Sr in component A (Asian dust, $(\text{Sr}) = 219.1$ ppm and $^{87}\text{Sr}/^{86}\text{Sr} = 0.71503$ (Pettke et al., 2000)), $(\text{Sr})_B$ and $(X)_B$ are the concentration and isotopic ratio of Sr in component B (volcanic glass from the Izu-Bonin Mariana arc system, $(\text{Sr}) = 183.9$ ppm, $^{87}\text{Sr}/^{86}\text{Sr} = 0.703208$ (Bloomer and Hawkins, 1987; Brounce et al., 2014; Fryer et al., 1982; Gribble et al., 1996, 1998; Hart et al., 1972; Hawkins et al., 1990; Ito and Stern, 1980; Lonsdale and Hawkins, 1985; Matthey et al., 1980; Pearce et al., 2005;

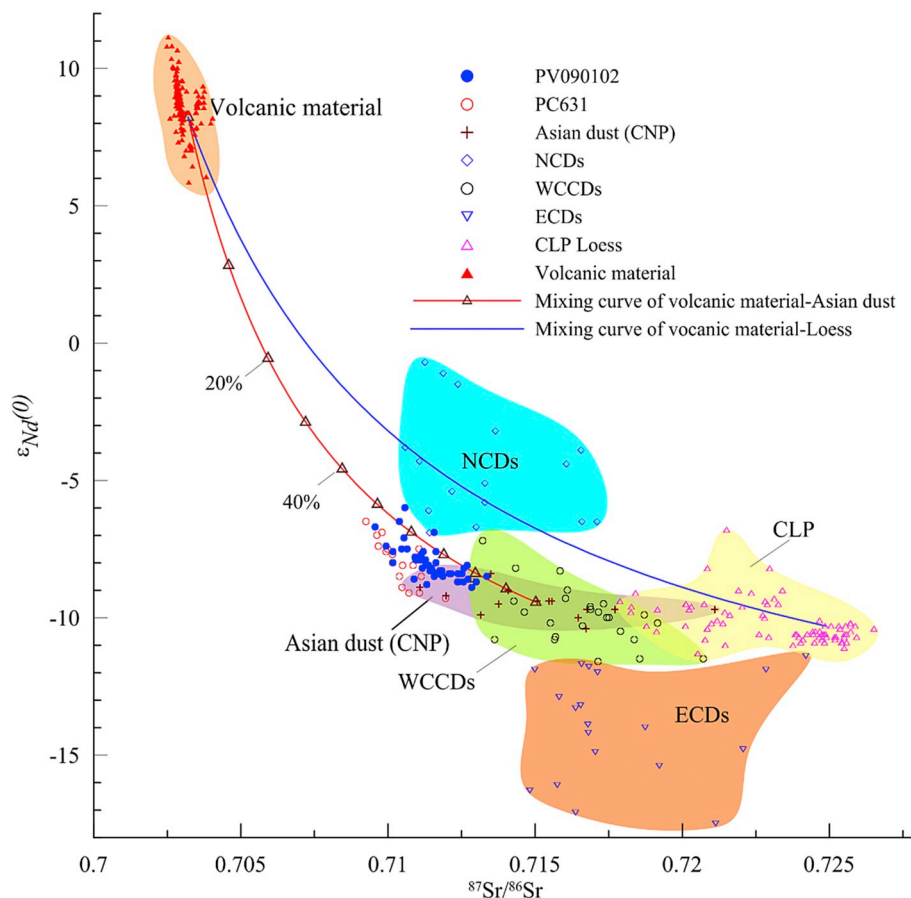


Fig. 5. $^{87}\text{Sr}/^{86}\text{Sr}$ vs $\epsilon_{\text{Nd}}(0)$ of the detrital fraction of the core PV090102 sediments, volcanic materials and potential dust source regions. “NCDs” refers to the northern Chinese deserts, including the Gurbantunggut Desert, Onqin Daga Sandy Land, Hunlun Buir Sandy Land and Horqin Sandy Land. “WCCDs” refers to the western and central Chinese deserts, including the Taklimakan, Qaidam, Tengger and Badain Jaran deserts. “ECDs” refers to the eastern Chinese deserts, including the Hobq and Mu Us deserts. NCDs, WCCDs and ECDs correspond to regions A, B and C, respectively, in Chen et al. (2007). “CLP” represents the Chinese Loess Plateau (Sun, 2005; Wang et al., 2007; Chen and Li, 2013). The red line is the mixing curve between volcanic glass from the Izu-Bonin Mariana Arc (Bloomer and Hawkins, 1987; Brounce et al., 2014; Fryer et al., 1982; Gribble et al., 1996, 1998; Hart et al., 1972; Hawkins et al., 1990; Ito and Stern, 1980; Lonsdale and Hawkins, 1985; Matthey et al., 1980; Pearce et al., 2005; Reagan and Meijer, 1984; Sinton and Fryer, 1987; Stern et al., 1991, 2013; Straub, 1995, 1997; Volpe et al., 1987, 1990) and Asian dust from the central North Pacific (CNP, Pettke et al., 2000), and the relative percentages of Asian dust are marked with empty triangles on the line, with each point indicating 10%. The blue line is the mixing curve between volcanic material and CLP loess. The mixing lines in the $^{87}\text{Sr}/^{86}\text{Sr}$ vs $\epsilon_{\text{Nd}}(0)$ plot were developed from known concentrations and isotopic signatures of Sr and Nd from CLP loess ($\text{Sr} = 98.7 \text{ ppm}$, $\text{Nd} = 18.8 \text{ ppm}$, $^{87}\text{Sr}/^{86}\text{Sr} = 0.724882$ and $^{143}\text{Nd}/^{144}\text{Nd} = 0.512102$) (Sun, 2005; Jahn et al., 2001; Chauvel et al., 2014), the Izu-Bonin-Mariana arc system, and Asian dust (see Fig. 3). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Reagan and Meijer, 1984; Sinton and Fryer, 1987; Stern et al., 1991, 2013; Straub, 1995, 1997; Volpe et al., 1987, 1990)), and $(X)_M$ is the measured Sr isotopic ratio in the detrital fraction of core PV090102. The calculated dust MAR results are shown in Table S3.

In the two end-member mixing calculations, we assume that (a) the Sr and Nd isotopic composition of the PVB, which ultimately reflects the eolian input from the Asian continent to the PVB, did not change significantly over the last 2.1 Myr; (b) the long-distance transported central North Pacific sediment represents the old continental crust end-member; and (c) the isotopic composition of volcanic glass from the Izu-Bonin Mariana arc system is representative of sedimentary input from island arcs to the PVB, and the isotopic composition is relatively stable. These assumptions are based on two important facts: (1) Previous studies have demonstrated that the Sr isotopic composition of volcanic materials is time independent in the western Pacific over the last 1.9 Myr (Ku et al., 2009). (2) The Sr isotope variations of volcanic materials and Asian dust in different time periods are relatively small compared with the large difference between volcanic material and Asian dust.

The eolian dust MARs over the last 2.1 Myr in core PV090102 are shown in Fig. 6 alongside MARs from other deep ocean records, such as in the central North Pacific (ODP site 885A, Rea et al., 1998) and western equatorial Pacific (ODP site 806, Patterson et al., 1999), and the Chinese loess record (Sun and An, 2005). The average dust MARs of core PV090102 are $44.6 \text{ mg cm}^{-2} \text{ kyr}^{-1}$, with a range from 17.8 to $67.9 \text{ mg cm}^{-2} \text{ kyr}^{-1}$. The estimated MARs of Asian dust in the PVB are similar to the MARs in the tropical North Pacific; for example, the estimated modern dust MAR on Bokandretok Island ($11^\circ 20' \text{N}$, $162^\circ 20' \text{E}$) from the Enewetak Atoll in the Marshall Islands is approximately

$50 \text{ mg cm}^{-2} \text{ kyr}^{-1}$ (Duce et al., 1980). However, the dust MARs in the PVB are significantly higher than those in the equatorial Pacific (e.g., ODP site 806 on the Onto Java Plateau, which has dust MARs that range from 3.8 to $15 \text{ mg cm}^{-2} \text{ kyr}^{-1}$; Patterson et al., 1999) and significantly lower than those in the central North Pacific (e.g., core V21-146 and ODP site 885A). The dust MARs in core V21-146 and ODP site 885A range from approximately $50 \text{ mg cm}^{-2} \text{ kyr}^{-1}$ to $> 500 \text{ mg cm}^{-2} \text{ kyr}^{-1}$ (Hovan et al., 1991) and from approximately 100 to $160 \text{ mg cm}^{-2} \text{ kyr}^{-1}$ (Rea et al., 1998), respectively. These latitudinal variations in dust MARs over the last 2.1 Myr are consistent with the regional distribution of dust MARs in the western Pacific (Rea, 1994), which shows that the highest Asian dust MARs are concentrated in a band that stretches eastward across the northwestern Pacific directly downwind from the Asian dust sources and decrease latitudinally. The variation in the dust MARs of core PV090102 can be divided into two periods (Fig. 6) over the last 2.1 Myr. From 2.1 Myr to 1.2 Myr, the dust MARs were relatively low and ranged from 17.8 to $32.5 \text{ mg cm}^{-2} \text{ kyr}^{-1}$, with an average of $27.7 \text{ mg cm}^{-2} \text{ kyr}^{-1}$. A gradual and small increase in dust MARs starting from 1.5 Myr can be observed during this time interval. During the last 1.2 Myr, the dust MARs doubled and ranged from 28.3 to $67.9 \text{ mg cm}^{-2} \text{ kyr}^{-1}$, with an average of $52.0 \text{ mg cm}^{-2} \text{ kyr}^{-1}$. The most dramatic rise in dust MARs occurred at approximately 1.0 Myr, when the MARs reached approximately $67.9 \text{ mg cm}^{-2} \text{ kyr}^{-1}$ (Fig. 6). The significant increase in the dust MARs (Fig. 6) in core PV090102 at approximately 1.2 Myr has also been documented in other deep-sea and land records (Fig. 6) (Rea et al., 1998; Patterson et al., 1999; Sun and An, 2005), which strongly support the notion that Asian dust was input into the PVB and critical environmental changes occurred at approximately 1.2 Myr.

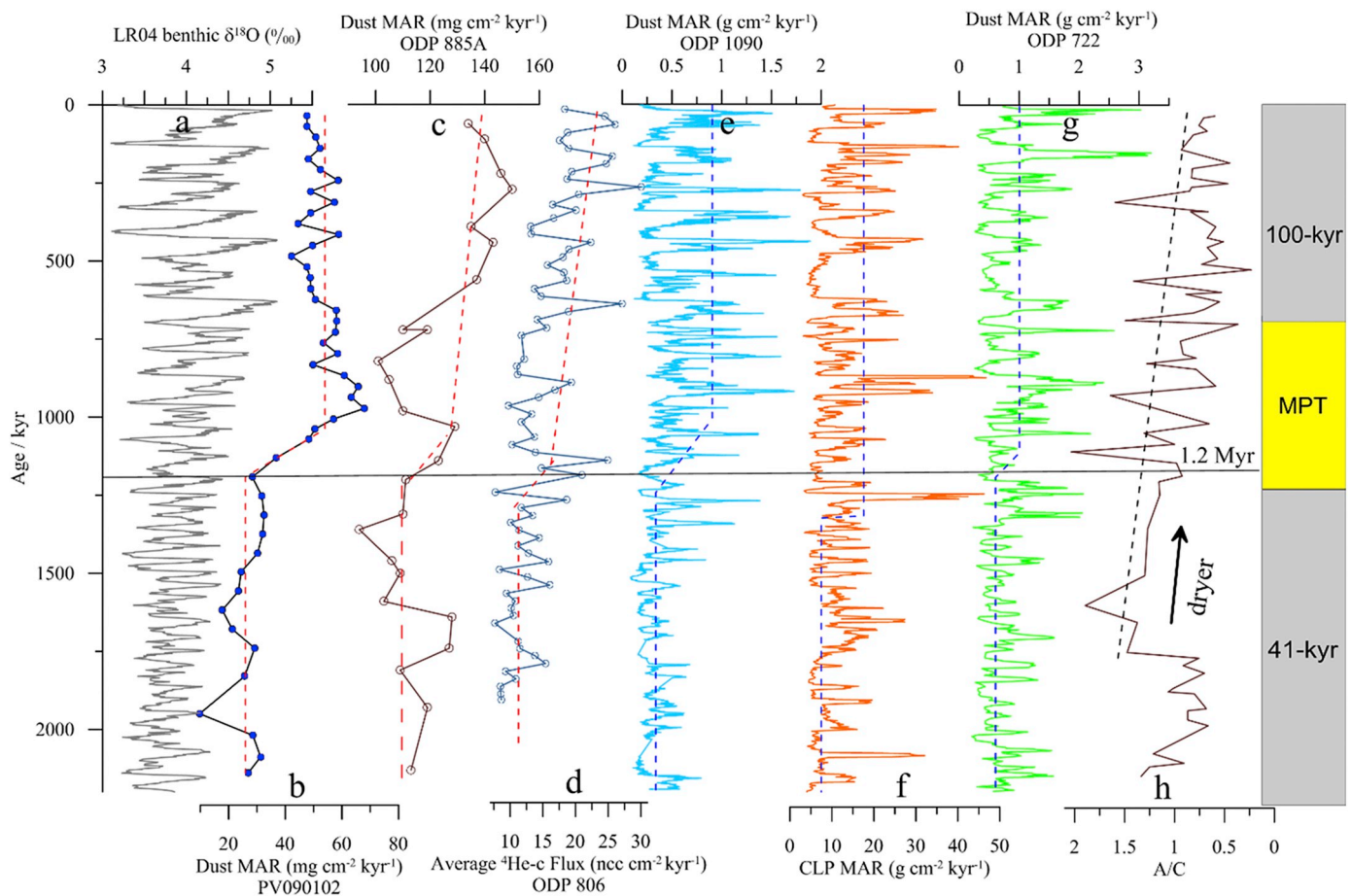


Fig. 6. MARs of dust in core PV090102 and other terrestrial and oceanic dust records compared to the benthic $\delta^{18}\text{O}$ record of ice volume change. (From left to right: a, the stacked benthic oxygen isotope record (Lisiecki and Raymo, 2005); b, dust MARs from core PV090102 (this study); c, dust MARs from ODP site 885A in the North Pacific (Rea et al., 1998); d, average $^4\text{He-c}$ flux from ODP site 806 in the western equatorial Pacific (Patterson et al., 1999); e, the MAR record from ODP site 1090 in the South Atlantic (Martínez-García et al., 2011); f, the mean MAR record from the central Chinese Loess Plateau (Sun and An, 2005); g, the MAR record from ODP site 722 in the Arabian Sea (Clemens et al., 1996); h, A/C is the ratio of *Artemisia/Chenopodiaceae* (Cai et al., 2012); the dashed lines indicate the variation trend, and the time interval of MPT is 1.25–0.7 Myr).

However, we noticed that the significant variation of dust MARs starting from 1.2 Myr is asynchronous with Sr and Nd isotope variation (Fig. 7) because the abruptly increased Sr and decreased Nd isotopes started at 1.5 Myr ago. As noted above, the detrital sediments in the PVB are mainly composed of Asian dust and volcanic material, and the variations in Sr and Nd isotopes were influenced by the relative contributions of the two end-members. The slight increase in dust MARs and decrease in volcanic material MARs starting at 1.5 Myr (Fig. 7) enlarged the relative contribution of the two end-members. In addition, due to the significant difference in Sr and Nd isotopic composition between evolved Asian dust and juvenile Pacific volcanic material (see 5.1), a minor increase in dust input may result in a major variation in Sr and Nd isotopes. Therefore, we argue that the considerable change in Sr and Nd isotopes at 1.5 Myr only indicates a minor Asian dust input. The significant increase in Asian dust input started at 1.2 Myr.

5.3. Response to the aridity of the East Asian continent

Aridification and atmospheric circulation models are two possible mechanisms influencing Asian dust supply. However, the fine grain size (average $3.2\mu\text{m}$), small grain size range ($3.0\text{--}3.4\mu\text{m}$), and gradually finer grain size (Fig. 7) during the early and mid-Pleistocene (Zhou et al., 2015) indicate no abrupt or substantial change in atmospheric circulation (e.g., westerlies). Therefore, we argue that the significant variation in dust input starting from the MPT was not driven by the

wind system.

The oxygen isotope records of deep-sea sediments (Lisiecki and Raymon, 2005) and dust MARs from ocean deposits (Rea et al., 1998; Clemens et al., 1996) have documented the long-term global trends of cooling and drying since the Miocene. Against this background, the Asian continent also experienced an evolution toward cooler and drier conditions (Sun and An, 2005; Rea et al., 1998). A series of stepwise aridification events at ca. 2.6 Myr, 1.2 Myr and 0.9 Myr were documented in sporopollen records from lacustrine-playa deposits in the western Qaidam Basin on the northeastern Tibetan Plateau (TP) (e.g., the *Artemisia/Chenopodiaceae* (A/C) ratio, Fig. 6; Cai et al., 2012) and Chinese Loess Plateau (Wu et al., 2004, 2007).

During the interval from 2.6 to 1.8 Myr, the Asian continent suffered rapid aridification, which was recorded by changes in vegetation in the Qaidam Basin (Wu et al., 2011; Cai et al., 2012), a transition from red clay to the loess-paleosol sequence on the Chinese Loess Plateau (Ding et al., 1999), and the eolian dust flux to the North Pacific (Rea et al., 1998). Although the limited age span (only extending to 2.1 Myr) and the low resolution for our record do not allow us to evaluate the phase relationship between the aridity and dust MARs, the aridity of East Asia during the early Pleistocene (Wu et al., 2011; Cai et al., 2012) was followed by an increase in dust input over the central North Pacific, equatorial Pacific and northwestern Pacific (Fig. 6). This consistent variation indicates that the dust MAR variation in our record was a response to the aridity of the Asian continent during this time interval.

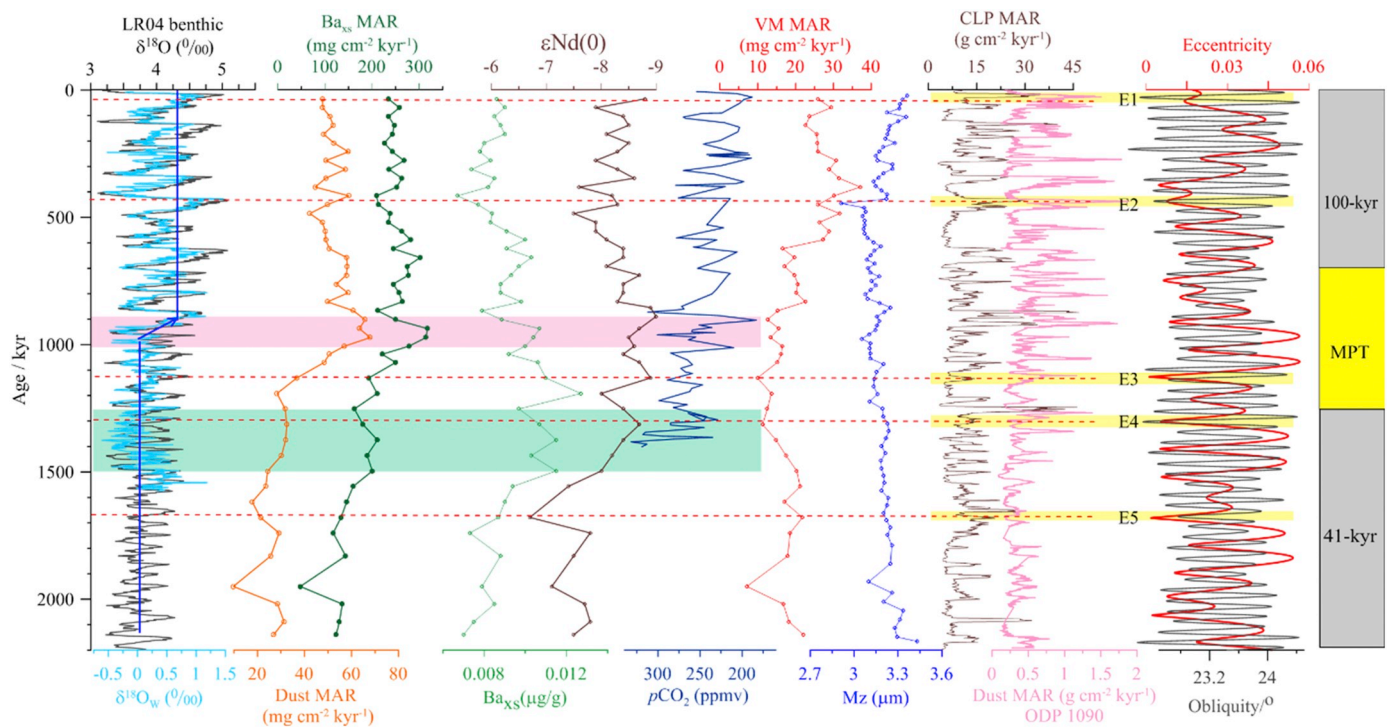


Fig. 7. Comparison of dust MAR, Ba_{xs} MARs, $\epsilon_{Nd}(0)$ and potential mechanisms of climate change. (LR04 Benthic $\delta^{18}O$ stack (Lisiecki and Raymo, 2005); $\delta^{18}O$ of seawater ($\delta^{18}O_w$, ice volume) (Elderfield et al., 2012); Mz (mean size of core PV090102) (Zhou et al., 2015) and pCO_2 (Tripathi et al., 2009). The CLP MAR is the dust MAR from the central Chinese Loess Plateau (Sun and An, 2005). Dust MAR of ODP site 1090 is from the South Atlantic (Martínez-García et al., 2011). Eccentricity and obliquity (Berger and Loutre, 1991); the sea green bar indicates the slight increase in dust MARs and decrease in volcanic material MARs starting at 1.5 Myr; the pink bar indicates the rapid decreased CO_2 concentration, significant increased dust MAR and export production at ca. 0.9–1.0 Myr; the yellow bars indicate the eccentricity and obliquity are in phase; the red dashed lines indicate the minimum eccentricity during E1, E2, E3, E4 and E5; the time interval of MPT is 1.25–0.7 Myr (Clark et al., 2006)). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Following this period, the climate in Asia experienced a short interval (from 1.8 to 1.2 Myr) of relatively moist conditions, as documented by vegetation from the northeastern TP (Fig. 6) (Cai et al., 2012). Responding to this wet climate, the dust contribution to the PVB, the central North Pacific and equatorial Pacific also decreased, as indicated by the relatively low dust MARs (Fig. 6).

Starting from ca. 1.2 to 1.1 Myr, the climate in the Asian interior became drier, as recorded by sporopollen proxies from the northeastern TP (e.g., A/C ratio, Fig. 6) (Cai et al., 2012) and Chinese Loess Plateau (Wu et al., 2004, 2007) and by the loess accumulation on the eastern TP (Yan et al., 2001). The stepwise advance of the Mu Us Desert recorded by eolian deposits in the desert-loess transitional area also supports the conclusion that the aridity in the dust source area occurred at approximately 1.1 Myr (Ding et al., 1999). The lower sand loess layer (L15) on the Chinese Loess Plateau, which likely resulted from an extremely dry climate, also appeared at 1.15 Myr (Liu, 1985). Sporopollen records on the Chinese Loess Plateau suggest a change from a forest-steppe to steppe environment at 0.95 Myr (Wu et al., 2007). This drying event is coeval with a large expansion of the Taklimakan Desert, as indicated by a strong expansion and grain size increase of loess sediments in the Tarim Basin at that time (Fang et al., 2002). The rapid increase in dust MARs from PV090102 beginning at ca. 1.2 Myr is nearly synchronous with the start of Asian aridification. The significantly higher average dust MARs during the period 1.0–0.9 Myr also matches well with the drying of the Asian continent (Fig. 6). Therefore, we argue that the dust MARs in the PVB over the last 2.1 Myr responded sensitively to the aridification of the Asian interior. This result is consistent with previous conclusions that the MAR of eolian dust to adjacent oceans is a proxy for the aridity of the dust source area (Rea, 1994; Tiedemann et al., 1994; Clemens et al., 1996).

5.4. Mechanism for increased dust MARs since the MPT

The most obvious feature of dust MARs in core PV090102 is the significant increase (from ca. $28.0 \text{ mg cm}^{-2} \text{ kyr}^{-1}$ to $> 60.0 \text{ mg cm}^{-2} \text{ kyr}^{-1}$) starting from ca. 1.2–1.0 Myr (Fig. 6). The increase in dust MARs is a global-scale change because it can be observed not only in the northwestern Pacific (e.g., core PV090102) but also in many other land and ocean records (Fig. 6), such as the Chinese loess record (from 10 to $30 \text{ g cm}^{-2} \text{ kyr}^{-1}$) (Sun and An, 2005), the central North Pacific (ODP site 885A; from 0.5 to $> 1.1 \text{ g cm}^{-2} \text{ kyr}^{-1}$) (Rea et al., 1998), western equatorial Pacific (ODP site 806; from 1.8 to $3.3 \text{ g cm}^{-2} \text{ kyr}^{-1}$) (Patterson et al., 1999), Arabian Sea (ODP site 722; from 500 to $1500 \text{ mg cm}^{-2} \text{ kyr}^{-1}$) (Clemens et al., 1996), South Atlantic (ODP site 1090; from 0.3 to $1.3 \text{ g cm}^{-2} \text{ kyr}^{-1}$) (Martínez-García et al., 2011), and equatorial Atlantic (ODP site 659; from ca. 0.5 to $> 1.1 \text{ g cm}^{-2} \text{ kyr}^{-1}$) (Tiedemann et al., 1994). As an indicator of the aridity of the dust source area, the dramatic increase in dust MARs indicates that the source region, such as east and central Asia, Africa and the Arabian Peninsula, experienced more dramatic aridity starting from approximately 1.2 Myr. This aridity is a global-scale climate change rather than a regional one. We argue that a global driving mechanism should be responsible for this abrupt increase in dust MARs.

Long-term global cooling has been recognized as a major global mechanism controlling the Late Cenozoic aridity history of Central Asia due to decreased water vapor held in the atmosphere (e.g., Lu et al., 2010; Miao et al., 2012; Sun and An, 2005). However, the global temperature cooling trend probably stalled before 1.2 Myr, and there is no direct evidence of gradual cooling over the MPT (Clark et al., 2006; Hönisch et al., 2009; Snyder, 2016). Moreover, no gradual increasing trend was observed in the dust MARs of core PV090102 over the last 1.2 Myr (Fig. 7). Therefore, other mechanisms that are not reliant on

global cooling seem more plausible for the abrupt and significant increase in dust MARs starting from 1.2 Myr in the northwestern Pacific.

The mid-Pleistocene transition (MPT), starting from approximately 1.2 Myr (Clark et al., 2006), represents a global evolutionary process in paleoclimate. During the MPT, the global climate shifted from a mode characterized by ~41-kyr cyclicity (obliquity) to a state dominated by ~100-kyr periods corresponding to eccentricity variations (Ruddiman et al., 1989). The global-scale processes and onset time are coupled with the global-scale increase in dust MARs and enhanced aridity (Fig. 6), which indicate that the driving mechanism of the global climate system transition during the mid-Pleistocene may have been responsible for the significant increase in dust MARs starting from 1.2 Myr.

The processes and mechanisms of the MPT remain in a state of persistent debate and conjecture (Clark et al., 2006; McClymont et al., 2013), although it is accepted that the external orbital forcing and/or internal feedback mechanism are responsible for the significant transition (Abe-Ouchi et al., 2013; Rial et al., 2013). The 41-kyr climate cycles during the Early Pleistocene is controlled by obliquity (Maslin and Ridgwell, 2005; Huybers, 2007), but the obliquity alone cannot explain the changes to the 100-kyr climate cycle during and after the MPT. Eccentricity affects insolation mainly by modulating the precession cycle; its direct contribution to the change in incoming insolation at glacial-interglacial timescales is too small to reconcile with the amplitude of the glacial cycles after 1.2 Myr (Raymo et al., 1997; Huybers and Wunsch, 2005; Berger et al., 2005; Lisiecki, 2010). However, the role of eccentricity is important in the early MPT large amplitude glacial-interglacial transitions (Clemens and Tiedemann, 1997) and becomes essential in combination with obliquity forcing.

The paced minimum eccentricity and low obliquity are very rare over the geological past and may trigger great climate change (Zachos et al., 2001). Comparing the eccentricity and obliquity (Berger and Loutre, 1991) reveals that the eccentricity minimum is totally in phase with low obliquity at time interval E4 and roughly in phase with low obliquity at E1, E2, E3 and E5 (Fig. 7). The frequency of the paced minimum eccentricity and obliquity obviously increased since MPT. The low resolution of our record does not allow us to establish a causal relationship between dust MARs and orbital variation; however, if we regard the MPT as a whole, the overall increased dust MAR since the MPT corresponds well with the frequent congruence of minimum eccentricity and low obliquity. In addition, as the dust MAR increase since the MPT is a global-scale process, we collected two high-resolution dust MAR records on the Chinese Loess Plateau (Sun and An, 2005) and South Atlantic (ODP site 1090) (Martínez-García et al., 2011) to compare with the eccentricity and obliquity variation (Fig. 7). The paced minimum of eccentricity and obliquity since the MPT matched well with or led the significant increase in dust MARs. In periods of low eccentricity, radiation exposure on Earth can accordingly fluctuate mildly between periods of perihelion and aphelion due to the nearly circular orbit. The planet's seasons are caused by the tilt of the Earth's axis. The low obliquity results in small temperature differences between summer and winter in the Northern Hemisphere. The combination of a minimum in eccentricity and a low-amplitude node in the obliquity cycle would reduce extremes of seasonal warmth and cold, allowing ice-sheet expansion and driving climate change (Zachos et al., 2001). Therefore, we argue that the increased congruence of minimum eccentricity and low obliquity triggered climate change before and during the MPT and drove the aridity of East Asia and the significant increase in dust MAR.

Although the external orbital forcing mechanism can explain the overall increased dust MARs since the MPT, the short period of congruence of minimum eccentricity and low obliquity cannot sustain the long-term increase of dust MARs (Fig. 7). An internal feedback mechanism is likely responsible for the long-term increase in dust MAR.

Dust has been demonstrated to play a key role in climate change by altering ocean biogeochemical cycles through changes in nutrients

(e.g., Fe carried by dust) and modifying atmospheric CO₂ concentrations (Martin, 1990). Evidence from the Southern Ocean shows a tight connection between increased dust and iron flux at the MPT (Martínez-García et al., 2011). A relationship between the giant diatom *E. rex* blooms in the PVB and enhanced flux of Asian dust during the Last Glacial Maximum has been suggested (Xiong et al., 2013, 2015). These diatom blooms sequestered atmospheric carbon and lowered atmospheric CO₂ during the glacial. Therefore, increased dust input potentially played a role in decreasing atmospheric CO₂.

In order to demonstrate the linkage between dust input and CO₂ concentration, a reliable proxy for paleo-productivity is required. Ba_{xs} is a widely used indicator of paleo-productivity in marine sediments, and a higher Ba_{xs} indicates a higher export production (Dymond et al., 1992; François et al., 1995). Xiong et al. (2012) have demonstrated that Ba_{xs} is a reliable proxy of export production in the PVB and have reconstructed the late Pleistocene export production variation based on the Ba/Ti ratio and Ba_{xs} (Xiong et al., 2012). Here, we use the same method to calculate the Ba_{xs} (Fig. 3) and reconstruct the Ba_{xs} MAR of the PVB sediment over the last 2.1 Myr (Fig. 7). The vertical variation of Ba_{xs} is similar to that of the Nd isotope (Fig. 7). The increased Ba_{xs} starting from 1.5 Myr was synchronous with the more evolved Sr and Nd isotopic composition (Fig. 7), and the high correlation coefficient (0.66) between Nd isotope and Ba_{xs} together indicates that the export production respond to the dust input (Fig. 7).

Over the last 2.1 Myr, the dust MAR and the Ba_{xs} in PV090102 have exhibited synchronous change (Fig. 7) and are highly related (correlation coefficient equal 0.86), indicating their causal relation. This evidence strongly supports the conclusion that the significantly increased Asian dust input since the MPT carried more nutrients to the PVB and fertilized the biology and stimulated the productivity increase. A comparison of the dust MAR in core PV090102 with the Ba_{xs} MAR and atmospheric CO₂ record (e.g., Tripathi et al., 2009) (Fig. 7) indicates that the significant increase in dust input was coupled with a significant increase in export production (high Ba_{xs} MAR) and a rapid decrease in atmospheric CO₂. The decreasing CO₂ corresponded to the gradually increased dust MAR and export production starting from 1.5 Myr. The rapidly decreased CO₂ corresponded to the significantly increased dust MAR and export production at ca. 0.9–1.0 Myr, all of which strongly supports the influence of dust input on the decreasing atmospheric CO₂. Studies have suggested that increased dust accumulation rates served as an internal positive feedback for the climatic reorganization during the MPT (Jickells et al., 2005). We argue that the increase in dust MARs in the PVB potentially resulted in the decrease in atmospheric CO₂ and may have played an important role in the aridification of the Asian continent, further resulting in greater dust formation and deposition since the MPT.

6. Conclusions

The ⁸⁷Sr/⁸⁶Sr and ¹⁴³Nd/¹⁴⁴Nd(*Nd*(0)) ratios of the detrital sediment fractions in core PV090102 from the PVB indicate the mixing of dust from the Asian continent and volcanic materials from the islands in and around the PVB over the last 2.1 Myr. Asian dust from the western and central Chinese deserts (WCCDs) is the major contributor to the evolved Sr and Nd isotopic signatures of the PVB's sediments.

A two end-member model of Sr isotope values was applied to quantitatively separate Asian dust from volcanic materials and to reconstruct dust MARs in core PV090102 over the last 2.1 Myr. The results indicate that dust MARs increased significantly starting at 1.2 Myr, which resulted from the enhancement of aridity across the East Asian continent. The significant increase in dust MARs was a global-scale change and was synchronous with the MPT. We argue that the congruence of the minimum eccentricity and the low obliquity may have triggered the climate change and resulted in the aridity of East Asia and increased dust input. The coupling between dust MAR and export production and atmospheric CO₂ strongly support the conclusion that

the internal positive feedback of eolian dust is an important mechanism resulting in greater dust input to the PVB since the MPT.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.gloplacha.2018.12.011>.

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